Stratospheric aerosol size distributions and optical depth: Long-term GEOS-Chem-UCX/APM simulations and comparisons with measurements

Fangqun Yu[1], Gan Luo[1], Arshad Arjunan Nair[1], and Jun Wang[2]

[1] Atmospheric Sciences Research Center, State University of New York at Albany
[2] Center for Global and Regional Environmental Research , University of Iowa

Acknowledgements: Data (GloSSAC (SAGE/ISS, CLIPSO), Balloon Data, Atom airborne measurements), GEOS-Chem support team, Funding Support from NASA

NASA SAGE III/ISS SCIENCE TEAM project (80NSSC21K1199)

"Sources and radiative forcing of upper tropospheric and lower stratospheric aerosols: an integrated analysis of the effects of pyrocumulonimbus events"

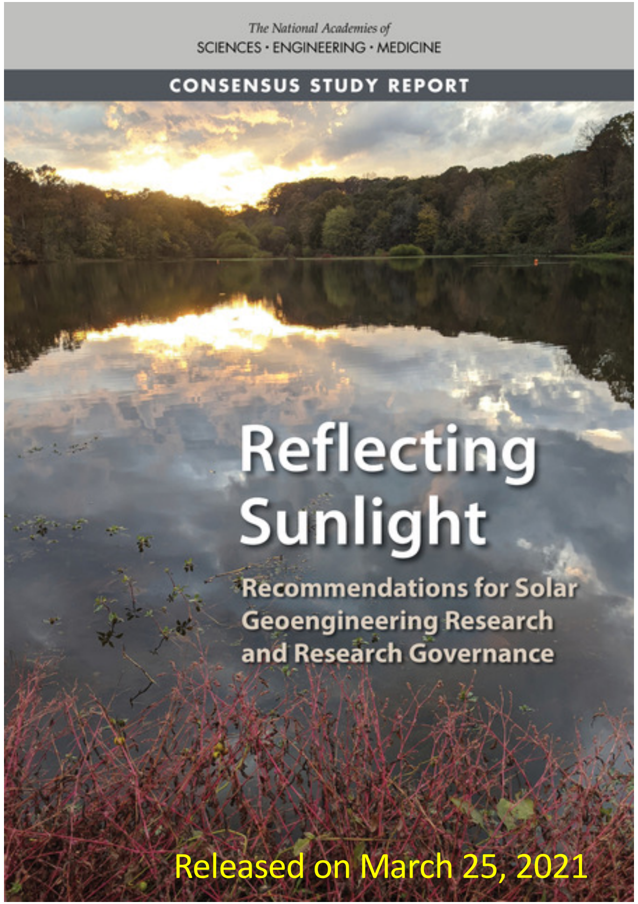
Principal Investigator: **Jun Wang**, University of Iowa

Co-Investigator: **Fangqun Yu** , State University of New York at Albany

One of research tasks: "**To simulate the evolution of size distribution of pyroCb particles and its possible coasting by sulfuric acid**"

Today's presentation focuses on particle size distributions of stratospheric particles <u>without</u> pyroCb

Understanding processes controlling particle size distributions in the stratosphere is important, especially for stratospheric aerosol injection (SAI)



Two quotes from the 2021 report by the National Academies of Sciences, Engineering and Medicine (NASEM)

"The overall magnitude and spatial distribution of the forcing produced by SAI depends strongly on the aerosol size distribution"

"One of the research priorities for SAI is thus to address critical gaps in knowledge about the evolution of the aerosol particle size distribution".

SAGE/ISS and other measurements of stratospheric aerosols are very useful to constrain our understanding.

GEOS-Chem-UCX/APM

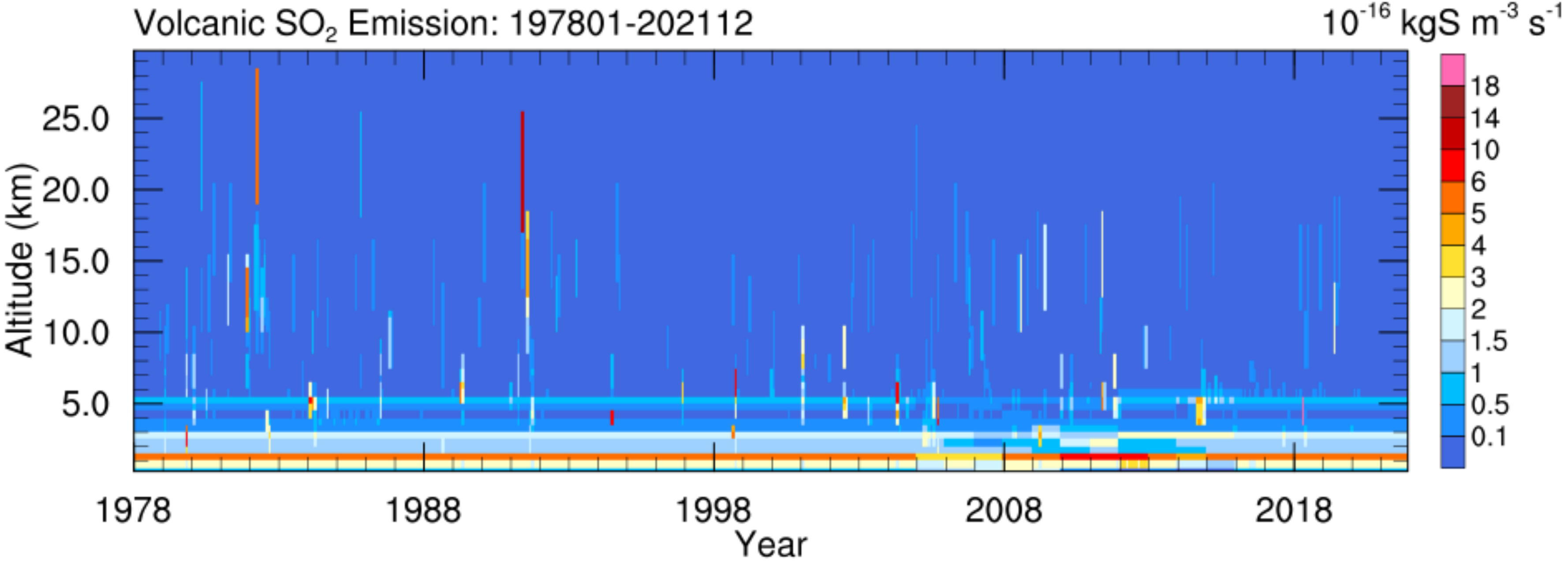
The GEOS-Chem **tropospheric–stratospheric unified chemistry extension** (UCX; Eastham et al., 2014), now the standard GEOS-Chem configuration, implements stratospheric chemistry, calculation of J-values for shorter wavelengths, and **improved modeling of high-altitude aerosols**. Extension of the chemistry mechanism to include reactions relevant to the stratosphere enables the capturing of stratospheric responses and **troposphere–stratosphere coupling**.

APM (Yu and Luo, ACP, 2009)

- **Sectional aerosol** tracers: Secondary Particles (40 bins), Sea Salt (20 bins), Dust (15 bins), BC (15 bins), POC (15 bins)
- **Coating** of SP on primary particles
- Nucleation, condensation/evaporation, coagulation, size resolved dry and wet deposition

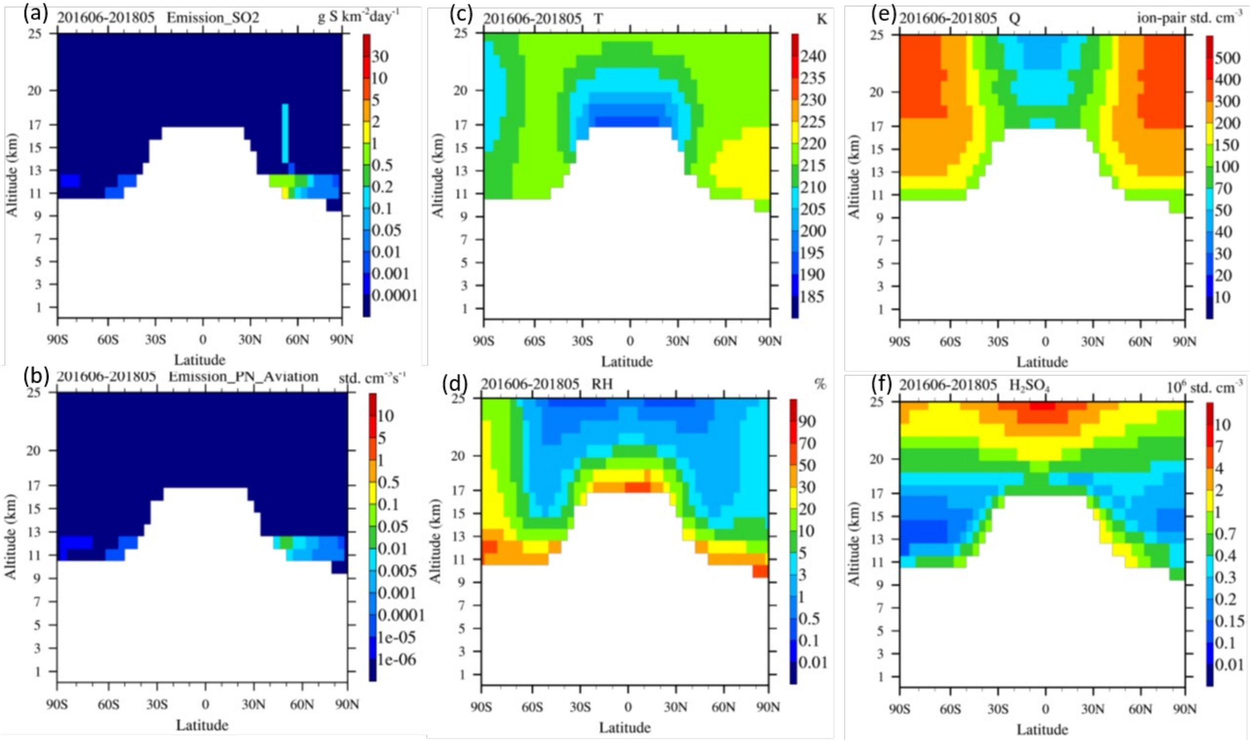
Long-term simulations: 1980-2021, 4^ox5^o, 72 vertical layers, MERRA2 met.

Up-to-date emission inventories (CEDS, NEI, APEI, GFED)
Volcanic SO_2 emission (Carn et al., 2015)



UV satellite measurements of volcanic SO_2 emissions by explosive and effusive eruptions from 1978 to 2021 based on Total Ozone Mapping Spectrometer (TOMS), Ozone Monitoring Instrument (OMI), and Ozone Mapping and Profiler Suite (OMPS) data

GEOS-Chem-UCX-APM simulation of PSDs in the stratosphere



Zonally mean SO$_2$ emission rate (a), direct emission of particle number due to aviation (b), temperature (c), relative humidity (d), cosmic ray ionization rate (e), and [H$_2$SO$_4$] (f) averaged during a two-year periods (201606-201805) covering Atom 1-4. To focus on lower stratosphere, only the values of these variables in the stratosphere (grid boxes with more than 50% time above tropopause) are shown.

Yu et al., ACPD, 2022

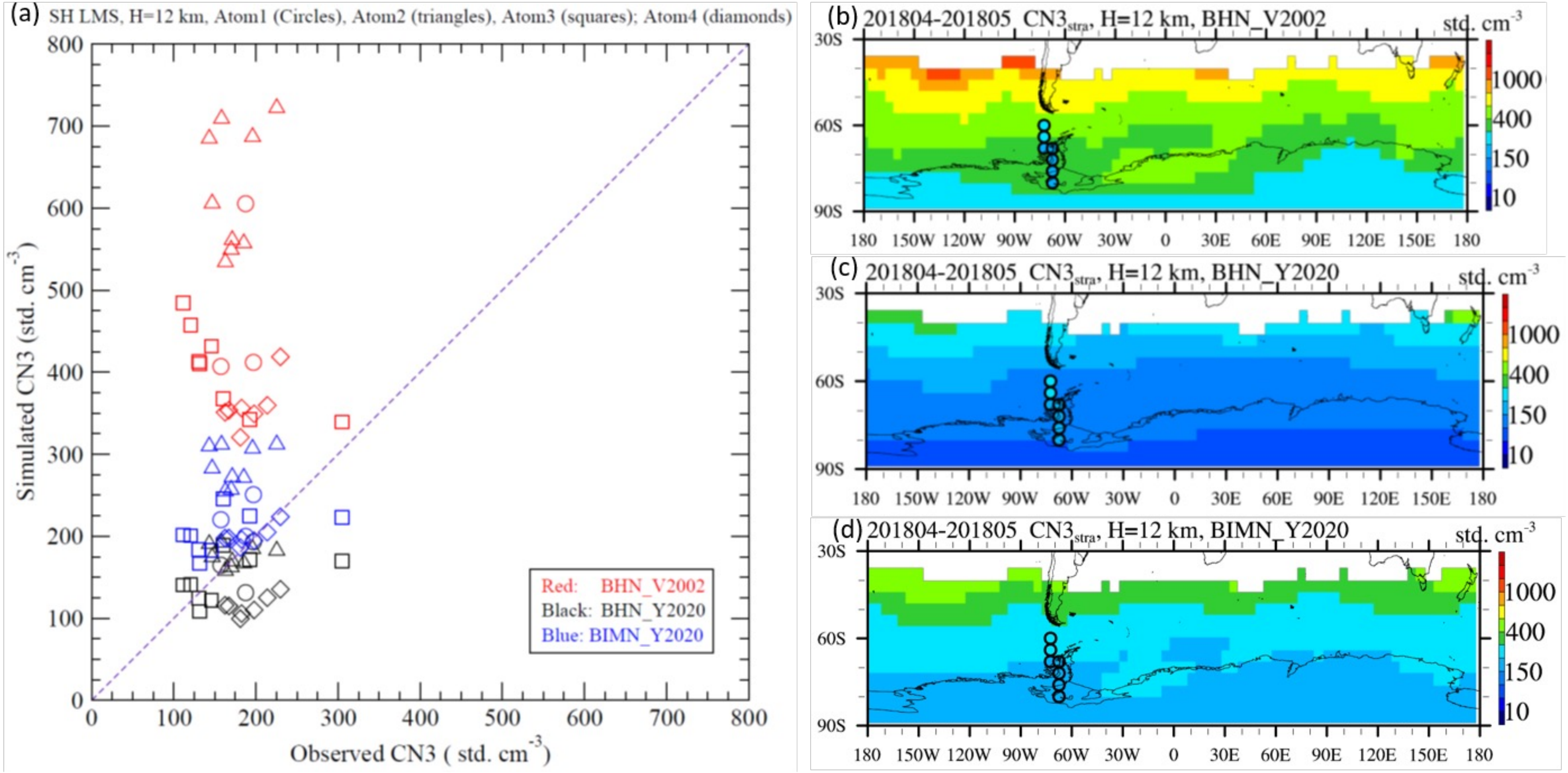
Nucleation schemes considered

(1) H_2SO_4–H_2O binary homogenous nucleation (BHN) scheme by Vehkamäki et al. (2002) (**BHN_V2002**),

(2) BHN of Yu et al. (2020) (**BHN_Y2020**), and

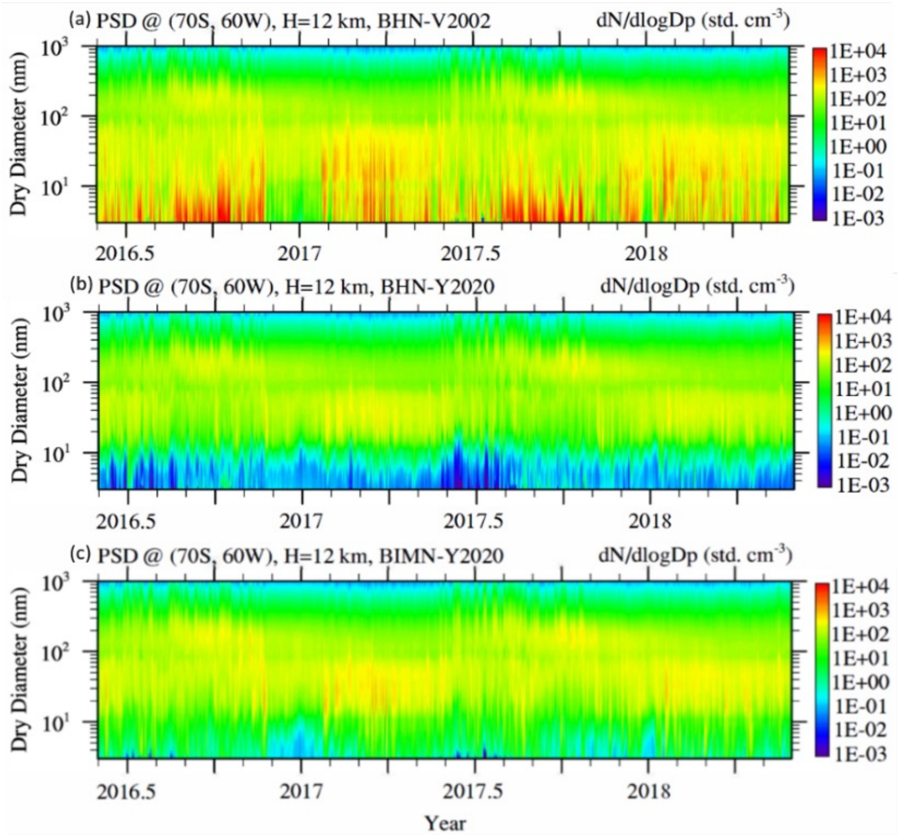
(3) Binary ion-mediated nucleation of Yu et al. (2020) (**BIMN_Y2020**)

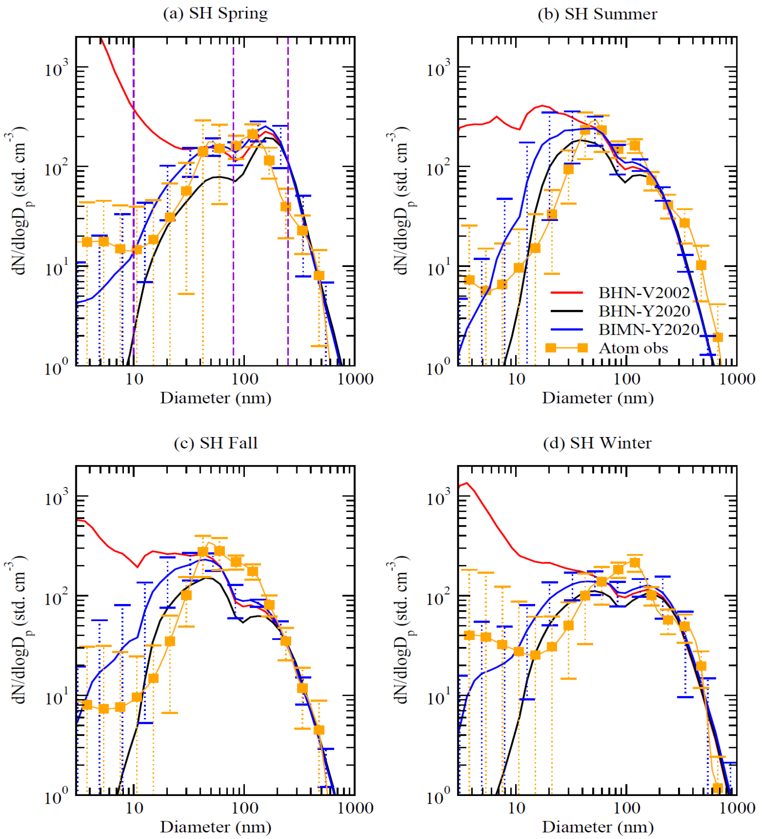
Comparisons with ATom measurements of condensation nuclei >3nm (CN3)



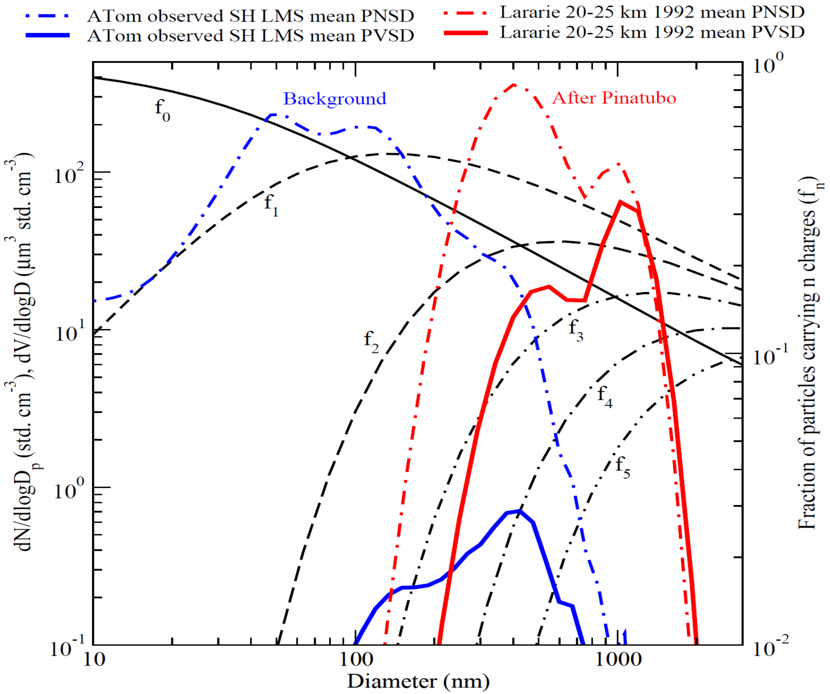
BHN_V2002 scheme overpredicted particle number concentrations in the background stratosphere by a factor ~2–4.

Comparisons with ATom PSD and implications





The model captures reasonably well the two modes (Aitken mode and the first accumulation mode) with the highest number concentrations and the size-dependent standard deviations. However, the model misses an apparent second accumulation mode peaking around 300–400 nm.



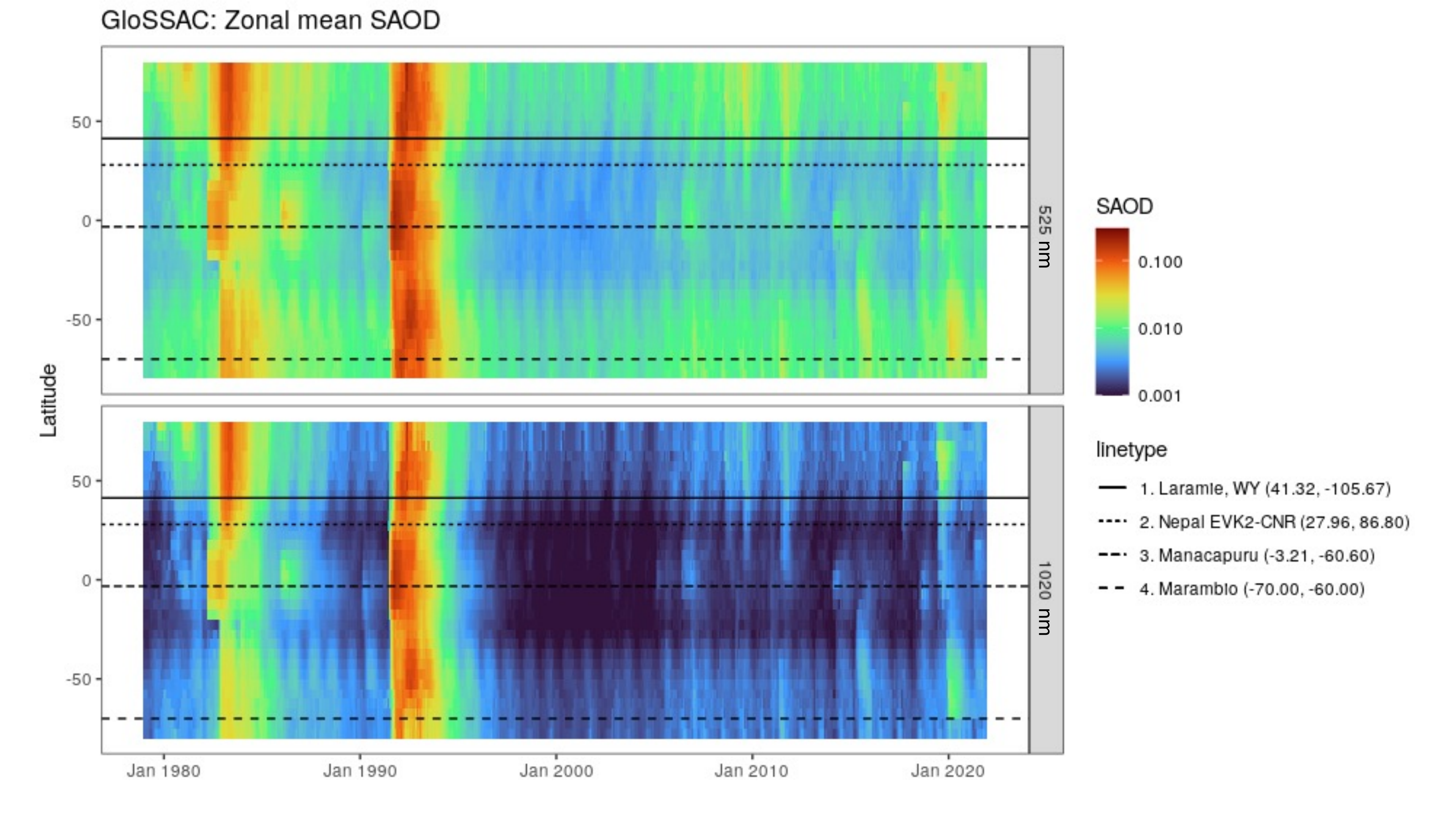
ATom 1-4 mean observed particle number size distribution (PNSD, or dN/dlogDp) and particle volume size distribution (PVSD, or dV/dlogDp) in SH LMS, balloon-borne measured mean PNSD and PVSD within 20-25 km altitude over Lararie WY in 1992, and fraction of particles carrying *n* (*n* = 0, 1, 2, 3, 4, and 5) charges based on the modified Boltzmann equilibrium equation (Clement and Harrison, 1992). Note that f_n with *n*≥1 including both positive and negative charges. (for details, see Yu et al., ACPD, 2022)

- The in-situ measurements indicate a bi-mode structure of accumulation mode particles both in the background stratosphere and in the volcano-perturbed stratosphere, which is not captured by the present model. The possible reasons remain to be studied.
- We suggest that the bi-mode structure may be caused by the effect of charges on coagulation and growth, which is not yet considered in existing models and may be important in the stratosphere due to high ionization rates and long lifetime of aerosols.

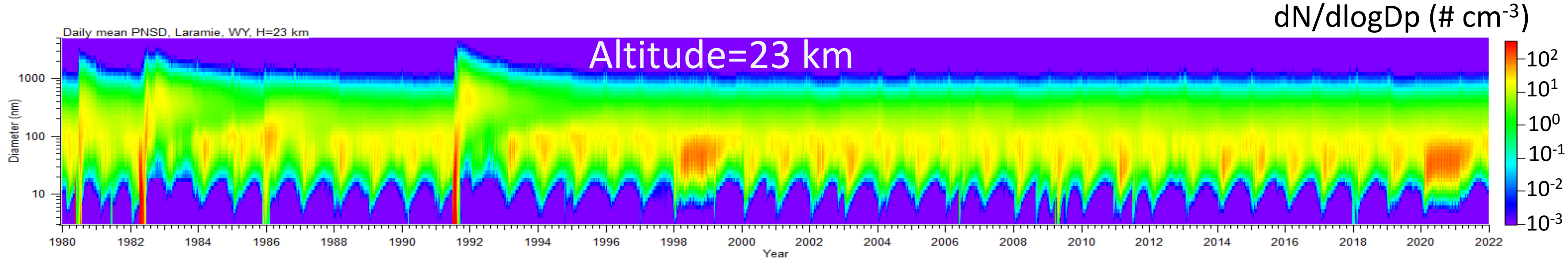
GloSSAC

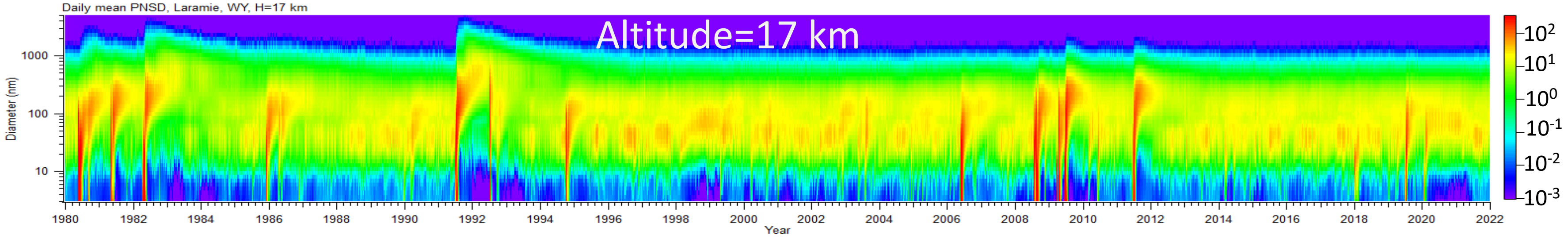
- **Global Satellite-based Stratospheric Aerosol Climatology**
- 43-year climatology of stratospheric aerosol properties focused on extinction coefficient (525 & 1020 nm)

Adapted from: Kovilakam, M., Thomason, L. W., Ernest, N., Rieger, L., Bourassa, A., and Millán, L.: The Global Space-based Stratospheric Aerosol Climatology (version 2.0): 1979–2018, Earth Syst. Sci. Data, 12, 2607–2634, https://doi.org/10.5194/essd-12-2607-2020, 2020.

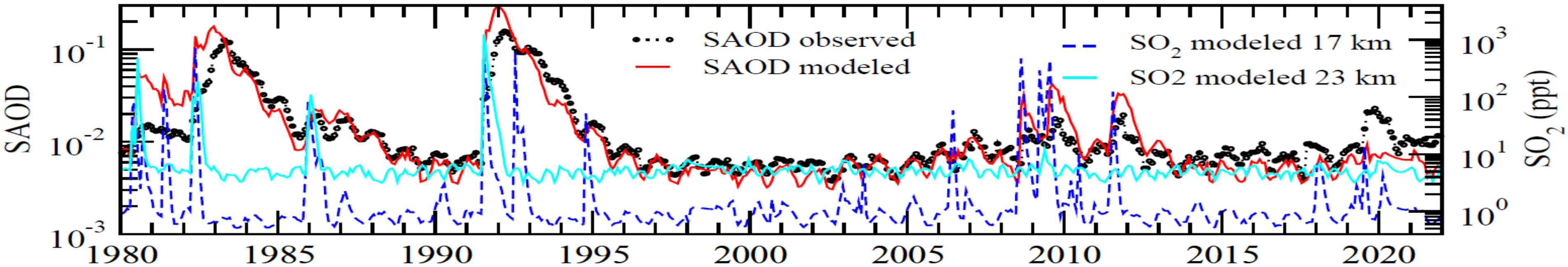

GloSSAC: Zonal mean SAOD
Latitude
50
0
-50
525 nm
1020 nm
Jan 1980
Jan 1990
Jan 2000
Jan 2010
Jan 2020
SAOD
0.100
0.010
0.001
linetype
1. Laramie, WY (41.32, -105.67)
2. Nepal EVK2-CNR (27.96, 86.80)
3. Manacapuru (-3.21, -60.60)
4. Marambio (-70.00, -60.00)

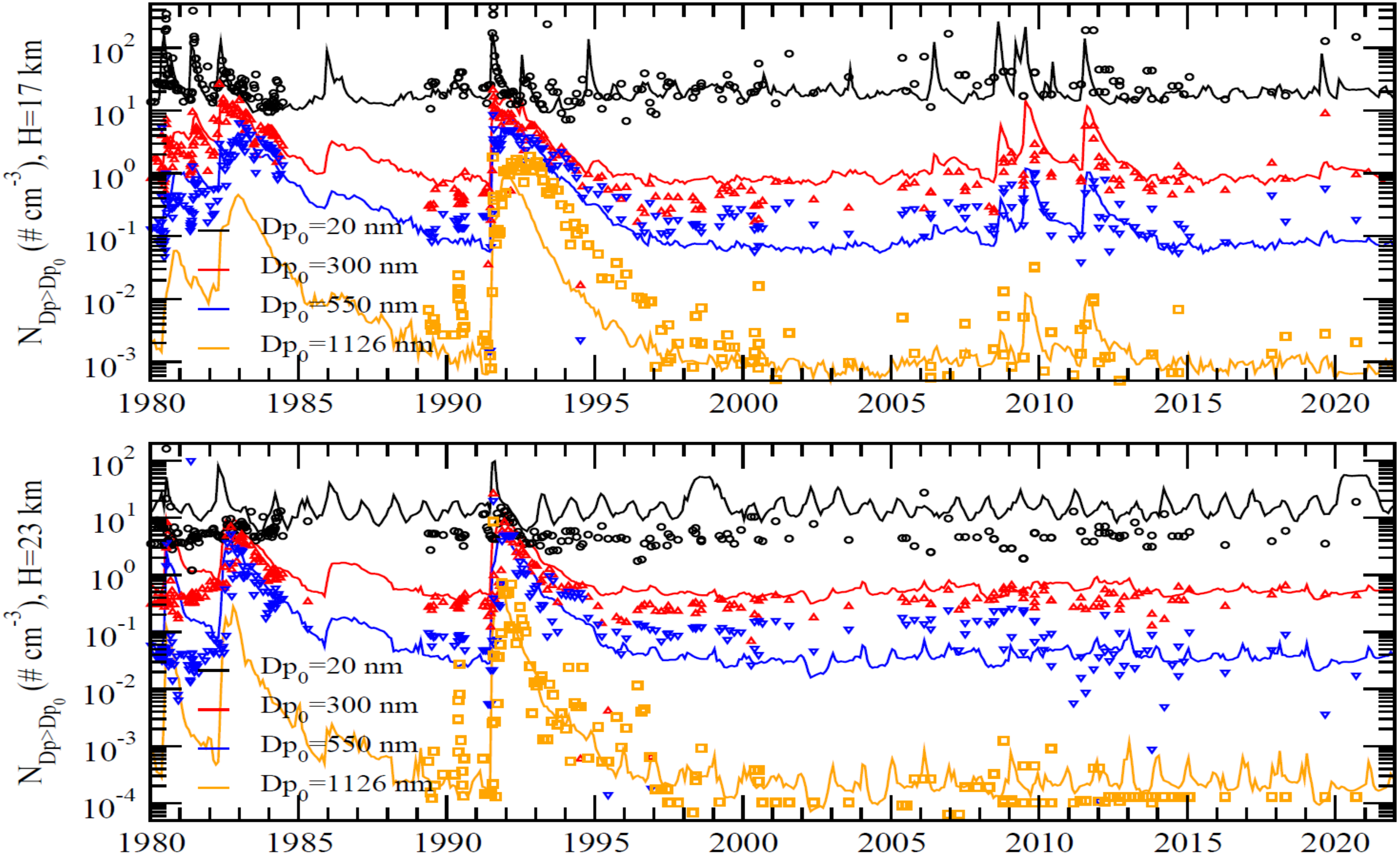
Daily mean particle number size distribution (PNSD), Laramie, WY





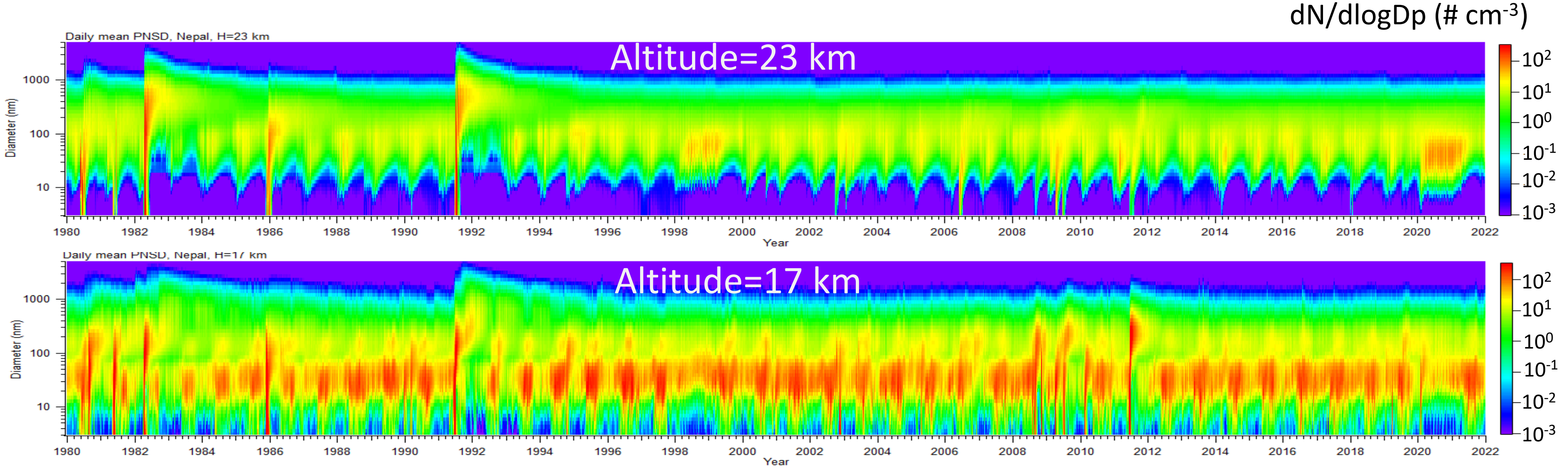
Lararie, WY, H=17 km and 23 km

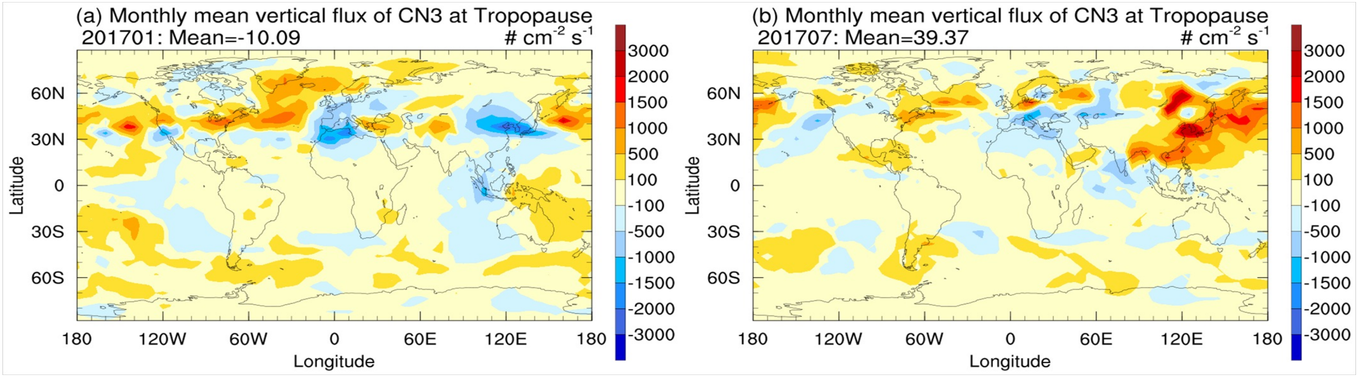




$N_{Dp>Dp_0}$ (# cm^{-3}), H=17 km
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-3}
Dp_0=20 nm
Dp_0=300 nm
Dp_0=550 nm
Dp_0=1126 nm
1980
1985
1990
1995
2000
2005
2010
2015
2020
$N_{Dp>Dp_0}$ (# cm^{-3}), H=23 km
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
Dp_0=20 nm
Dp_0=300 nm
Dp_0=550 nm
Dp_0=1126 nm
1980
1985
1990
1995
2000
2005
2010
2015
2020

Daily mean particle number size distribution (PNSD), Nepal





Strong upward transport of particles at the tropopause over Asia can be seen in summer

Summary

Questions? Please contact fyu@albany.edu

- APM has been integrated with UCX and the resulting GEOS-Chem-UCX-APM has been used to study the evolution of particle size distributions in the stratosphere.
- Nucleation schemes have a strong effect on the model simulated particle number concentration and size distribution in the stratosphere.
- The in-situ measurements indicate a bi-mode structure of accumulation mode particles both in the background stratosphere and in the volcano-perturbed stratosphere, which is not captured by the present model. The possible reasons remain to be studied. The effect of charges on coagulation and growth of particle in the stratosphere may be one of reasons.
- Comparisons of long-term simulated SAOD with GloSSAC data and balloon data show that the model generally capture the observed variations but indicate some differences, which remains to be further investigated.
- The Asian tropopause aerosol layer (ATAL) associated with Asian monsoon anticyclone appears to be a significant sources of stratospheric particles.